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Studies of the interaction of slow very highly charged ions with solid surfaces using the LLNL EBIT facility

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A summary of the present status of the LLNL EBIT based research program on highly charged ion–solid surface interaction is presented. New results for coincidence measurements of K^{α}–K^{β} X-ray emission for Fe^{26+} incident on Al, using two Ge detectors are presented. It is shown that excellent resolution of the satellite and hypersatellite lines are achieved, in excess of what is possible with single Ge detector measurements. Recent results from ongoing investigations on electron emission, sputtered ion and energy loss phenomena are presented. Results from charge-state analysis of grazing incidence specular scattering of highly charged ions off atomically flat surfaces of gold, carbon and mica indicate that the incident ion essentially reaches ground state of its electronic distribution within the ion–surface interaction time of ~100 fs. Atomic force microscopy is used to investigate the formation of a new type of ion induced surface defect, peculiar to highly charged ion–surface interactions. A linear correlation, between the defect volume and the incident ion charge, supports a mechanism of defect formation due to potential energy induced electron emission and subsequent collective displacement due to local charge imbalance.

1. Introduction

The interaction of highly charged ions (HCI) with solid surfaces is a relatively new topic within the field of ion–solid interactions and has received considerable interest in the last few years [1–5]. This is due, to a large extent, to the availability of HCI beams with sufficiently high flux, as produced by sophisticated ion sources such as an ECR [6] (electron cyclotron resonance) and EBIS [7] (electron beam ion source) and an EBIS variant, the EBIT [8,9] (electron beam ion trap). It is the use of the latter which is the topic of this report. The EBIT has been operating as a source by means of an efficient ion extraction system, which allows the execution of ion collision experiments with slow ion beams ($2.5 \leq E_{ext} \leq 15$ keVq) with very high charge states, q, up to U^{80+}. These ions have a total potential energy of about 200 keV, with associated fields of up to 10^8 V/cm. The physical concept of the EBIT is described in detail in ref. [8] and the use of EBIT as a source is described in ref. [9]. The available flux of few-electron high Z ions (such as Fe^{26+}, Kr^{36+}, Xe^{50+} and Th^{75+}) that can be extracted from the EBIT are sufficient to study e.g. electron emission, X-ray emission, surface sputtering and ion scattering, charge exchange, energy loss and surface defect formation, all of which are part of the EBIT HCI-surface interaction program. These allow the study of the mechanisms by which the HCI loses its excess potential energy when interacting with the surface, which is of fundamental interest. The spectroscopy of emitted X-rays and electrons during this interaction are the two most common methods established in this field for studying the various processes which are involved in the dynamics of the neutralization and re-establishment of equilibrium of the ion's electronic structure, as it approaches and penetrates the surface. It is currently understood that slow ions capture electrons efficiently into very high n or continuum states at relatively large distances from the surface, depending on the ionic charge [10–12]. This leads to the formation of a highly exotic excited state of the ion, where the electrons occupy high Rydberg levels while the core is virtually empty. It is the study of the formation and decay of these "hollow atoms" that has recently been the subject of intense investigations (e.g. refs. [4,5,13–16]). However, even quite sophisticated measurements of X-ray [5] and electron emission [13–15] have so far proven insufficient in providing a complete understanding of these processes. For instance, whereas the

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surface of a conductor may provide an essentially limitless source of electrons during the neutralization process, as indicated by recent electron emission studies [14], the limited density and mobility of nearly free electrons in insulators has recently been found to cause the production of nanometer sized defects. Recent results of such defect production studies, as well as a summary of the present status of our research program, are reported here, acting as an introduction of this facility to the atomic collisions in solids community. As it is clearly impossible to adequately address all of the issues involved within this short paper, the reader is necessarily referred to the literature, including a recent review of previous results using the EBIT facility [17] for further discussions of these topics.

2. Experimental setup

A schematic of the EBIT source and the extracted ion optics is shown in Fig. 1. The production and extraction of HCIs in this facility have been described in refs. [8,9]. The lower part of this figure displays nominal charge state distributions for extracted Th and U ions. Depending on the ion species and charge states, extracted ion rates between 10^4 and 10^6/s are obtained. Typical beam spots on the target are 2–3 mm in diameter and the emittance is less than π mm mrad. The residual gas pressure in the transport system was $\lesssim 2 \times 10^{-8}$ Torr, which causes charge exchange of the ions of less than 20%. The vacuum in the target chamber currently ranges from 5×10^{-10} to 2×10^{-8} Torr, depending upon the experiment. The arrangement of the sample depends upon the particular investigation which are briefly described below.

3. Electron emission studies

The energy distribution and yield [18], as well as the statistics [14] of electron emission, due to the impact of HCI on solid surfaces, have been measured for ion charge states up to Th^{80+} using EBIT. It has been shown that the emission yield is essentially linear with the incident charge over the entire range from 20 + to 80 +. Furthermore, additional information concerning the velocity limit, imposed by the effect of the ion's image charge, has been extracted from the yield–velocity relationship for critically decelerated incident ions [14]. The latter study has been accomplished as part of a collaboration with the group of Winter and Aumayr et al. of Vienna. More details of these results are presented elsewhere in these Proceedings by Aumayr et al.

4. X-ray emission studies

Briand et al. [5,19] has previously shown that high resolution measurements of the emitted X-ray energy

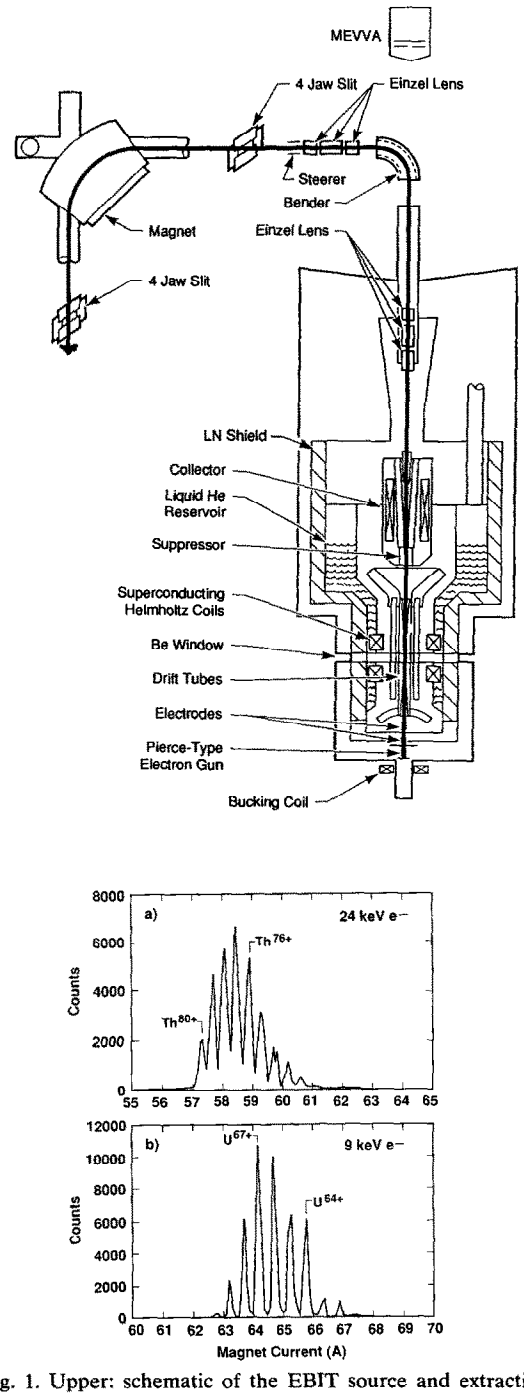

Fig. 1. Upper: schematic of the EBIT source and extraction system. Lower: nominal extracted charge state distributions for U and Th ions.

distribution in HCI–surface collisions can be used to determine upper limits on the transition rates which govern the collapse of the hollow atoms. This technique is based upon the determination of the time evolution of the number of spectator electrons between the emission of the hypersatellite and satellite lines and requires a crystal spectrometer in the case of Ar [5]. However, whereas the KL^x satellites are more widely separated in the case of Fe and Kr ions, a Ge detector can be used to study these higher charge state systems, by analyzing the moments of the X-ray energy distribution. In this way, the average number of L spectator electrons at the two steps of the K shell filling for both Fe^{26+} and Kr^{36+} were measured using EBIT, as part of a collaboration with the group of Briand et al. of Paris [20]. The resolution of the detectors was ca. 210 eV. The results indicate that the number of additional electrons in the L shell, Δn, during the emission of the hypersatellite and satellite X-rays, decreases with increasing Z of the incident ion (from $\Delta n = 4$ for Ar to $\Delta n = 2.5$ for Fe and $\Delta n = 1$ for Kr, not accounting for the "active" electron which filled the K-shell). The corresponding K shell vacancy lifetimes are 2 (Ar), 1 (Fe) and 0.4 (Kr) fs. These results have been interpreted, with the assistance of Dirac–Fock calculations, as suggesting that capture processes, just beneath the solid surface, directly fill outer shells with higher n number for Fe and Kr than for Ar. In an effort to gain more detailed information on the dynamics of these transition rates and as a continuation of this successful collaboration, we have performed an investigation of the correlated emission of the two K X-rays for 162 keV Fe^{26+} normally incident on an Al target. Two Ge detectors were arranged to provide a large detector solid angle (near π sr). A plot of the correlation results is shown in Fig. 2. It can be seen that the satellite and hypersatellite lines are better resolved than would be possible with a single Ge detector and that even higher order transitions are well resolved. These results allow an estimate for the dependence of the transition rates on the condition of the inner shell occupancies. Analysis of this data is under way.

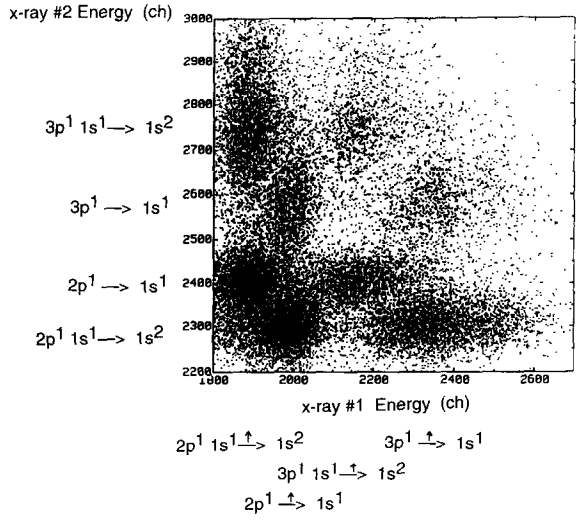

Fig. 2. Correlation plot of the K X-rays produced through the filling of the K shell in bare Fe^{26+} incident on an AlO target. Transition assignments to the measured lines are noted.

5. Sputter yield measurements

As sputtering cascades are usually generated below the surface of the solid, it is particularly interesting to determine if there are any significant effects of the incident charge state on the sputter yield. This would provide information of the electronic structure of the ion inside the solid, a regime not directly accessible to electron or X-ray emission techniques. Results of earlier studies of the effect of the incident charge state of multiply charged ions (up to $q = 9$) [21] on sputter yields suggest an effect on the charged but not on the neutral component, which was three orders of magnitude larger. As it may be thought, that long range coulomb interactions or changes in short range screening strengths, due to the charge of the incident ion, would increase the total sputter yield in insulators [22], further experiments involving higher charge ions were suggested. Preliminary experiments have been performed, in collaboration with Schiewitz of Berlin, using a time-of-flight system to measure the charged fraction of the sputter yield for SiO_2 and Xe^{q+} for q from 30 to 50 + [23]. It has been demonstrated that the yield of the positively charged component of the sputter/desorption yield, as well as the photon and electron yields increase with increasing incident charge states. Most recently, it has been found that the negatively charged component of the sputter/desorption yield also increases with increasing incident charge state and is comparable in intensity with the positively charged component. Additional studies, directed toward determination of the total target sputter yield, have been initiated in a joint collaboration with Biersack of Berlin and Cahill of California. The main difficulty in the later study is the extremely low fluence which is achieved for slow few-electron high Z ions of some 10^{10} cm^{-2}, necessitating highly sensitive analytic techniques to measure the sputter yield (such as PIXE).

6. Energy loss measurements

As in the case of total sputter yield, it has been suggested that additional energy loss due to long range

Coulomb interactions of HCI in insulating solids should be significant [22]. Such effects would also be of interest in the investigation of the transient charge of the HCI inside solids. In a further collaboration with Biersack, projected range measurements, for Xe^{q+} with $q = 1$ to 44 +, have been performed [24] in SiO_2 targets and incident energies of 110 and 300 keV. Again, the low HCI fluence requires the development of sensitive measurement techniques. In this case, the range of the ions was measured using heavy ion (2.4 MeV Ne +) Rutherford backscattering and a 16 segment detector array arranged to provide a solid angle of 0.15 sr. The detection limit of the setup is about 5×10^{10} Xe cm^{-2} in SiO_2 on Si. Details of the measurements have recently been presented elsewhere [25]. Further investigations involving transmission energy loss measurements with various insulating targets are in progress.

7. Small angle scattering

The use of small angle scattering of HCIs, off atomically flat crystal surfaces to study the transient ion–surface interaction has been established experimentally by Winter et al. [16] and Meyer et al. [26] and theoretically by Burgdorfer et al. [4]. Typically, either the angular distribution and image charge acceleration [16], the Auger electron yield [26] or the final charge state distribution [21] have been determined. Generally, these studies have been limited to incident charge states of $\leq 10+$. There has yet to be a simultaneous determination of the angular distribution, energy loss and charge-state distribution of the scattered ion, and little data at all for the highest charged high Z ions, where sidefilling of the inner shells during impact is expected to be minimal for the impact parameters associated with specular reflection. Such measurements have been performed using EBIT for Kr^{q+} ions with $q = 14$ to 35 on flat surfaces of mica, highly oriented pyrolytic graphite (HOPG, C(0001)), and epitaxially grown Au (111) on mica. Macroscopically defined incident angles varied between 0.05 and 2.5°, though image charge effects increase the actual incident angle to 1.5 to 5.5°. Energy losses are less than 0.5% and indicate large impact parameter, low dispersion collisions, as supported by clear specular reflection peaks in the scattered ion angular distribution. Results indicate that most ($\geq 70\%$) of the scattered ions are either neutral or singly charged, suggesting that the inner shells of the ion are essentially filled, outside the solid, within the interaction time of less than about 100 fs. This supports the extremely fast transition rates obtained from the X-ray emission data. Furthermore, a strong influence of the incident velocity, as well as the initial charge state has been found for the ratio of charged to neutral scattered ions. It has been found that the charged to neutral ratio of the scattered ions, for a given incident ion, decreases from Au to C to mica, indicating a stronger interaction for the insulator compared to the metal. This is supported by results of measurements of the energy gain of the ions due to image charge effects, applying a modified version of the method of Winter [16], using the shift in the angular distribution of specularly scattered ions as a function of the incident angle. These results indicate a significant increase in the energy gain, for a given incident ion, from Au to C to mica. Furthermore, it has been found that a significant yield (30–50%) of negatively charged scattered ions is produced for incident O^{7+}. This also increases from Au to C to mica. Details of these measurements are still being analyzed and a complete report will be presented shortly.

8. A new surface defect

Using ions produced by EBIT, in combination with atomic force microscopy (AFM), it has recently been established [27] that a new type of defect is produced through the impact of slow very HCI on insulating surfaces. The defect on mica is characterized by a hillock structure some 5–100 nm in diameter and 0.3 to 2 nm in height and is not formed for singly charged heavy ions at the same velocity of 2.2 keV/u. AFM results for such a defect, formed by U^{70+} impact on mica, is shown in Fig. 3. Whereas it has been found possible (but not normal) to peel these hillocks off, exhibiting a pit beneath, they have been designated "blisters". The exact mechanism of the blister formation is not clear, though it has been proposed that for mica, charge imbalance between the uppermost layers, due to substantial electron emission, cause the layers to dissociate locally. It should be noted that similar defect structures have been seen on Lexan, which has no layered structure. Very recently, a systematic study of the relationship between the defect volume and the incident charge state of the ion has been performed. A population distribution for defect volumes for Xe^{50+} on mica is shown in Fig. 4. It can be seen that there is significant variation in the measured defect volume. This variation is believed to be related to the statistics of the HCI–surface neutralization process [14], though investigations into possible intrinsic variations in the AFM results are currently proceeding. Fig. 5 shows the measured defect volume vs. incident charge for $q =$ Kr^{35+} to Th^{74+}. As can be seen, the defect volume is linearly proportional to the incident ion charge, exhibiting an apparent threshold at ca. $q = 28+$. This behavior is consistent with the formation of defects based on ionization density, as in nuclear track forma-

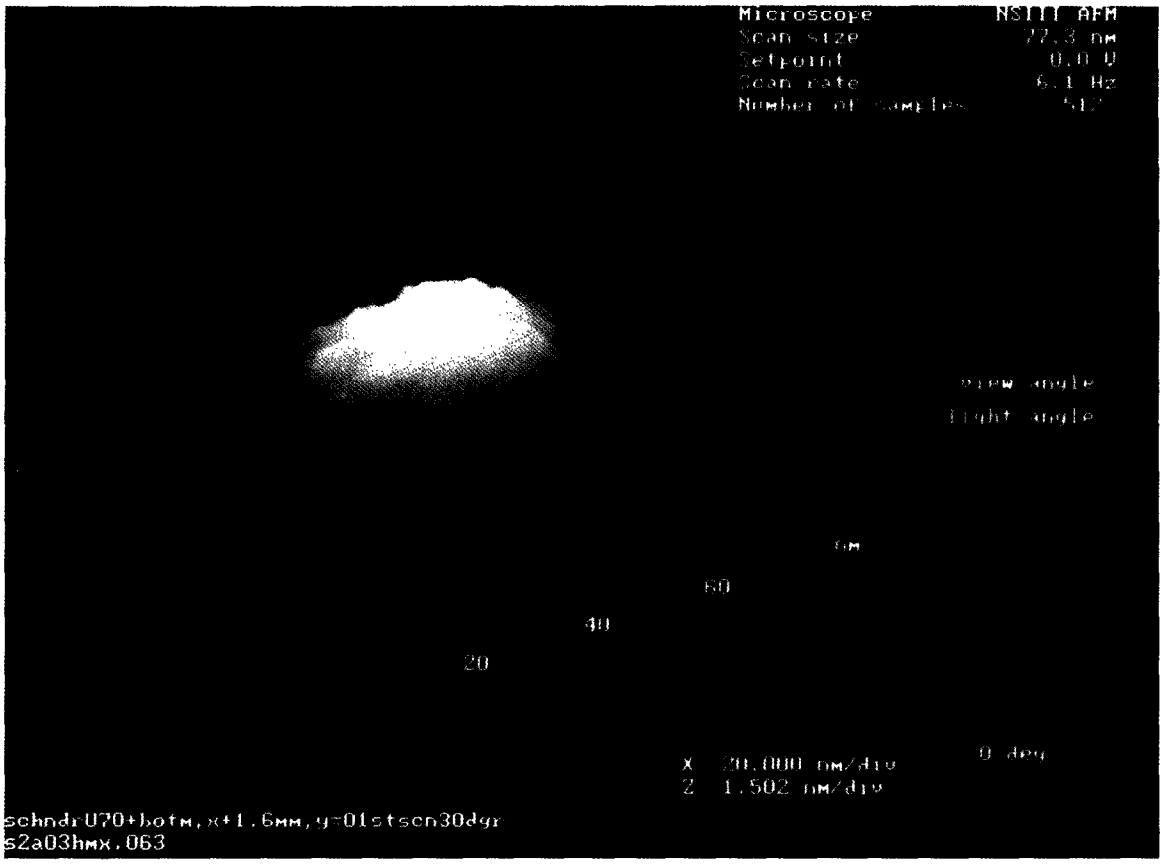

Fig. 3. Atomic force microscopy topographic reconstruction of a "blister" type surface defect on mica, caused by the impact of a single U^{70+} ion at 490 keV.

tion. In the present case, the ionization is due to potential energy induced emission, whereas the nuclear track formation is based on kinetic energy transfer to the target electrons. Further investigations into nanometer size surface defects may be useful in new generations of lithographically defined devices.

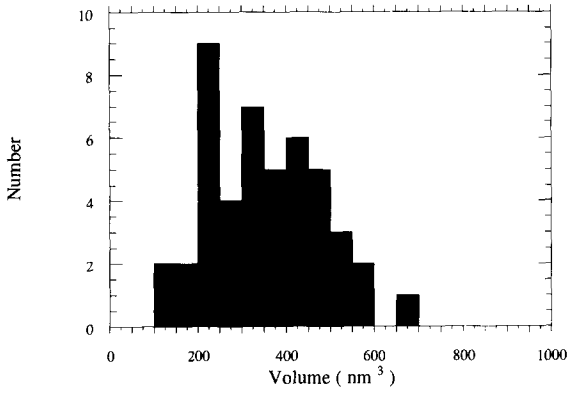

Fig. 4. The population distribution of measured volumes of blisterlike defects, determined through an atomic force microscopy, due to the impact of Xe^{50+} on mica.

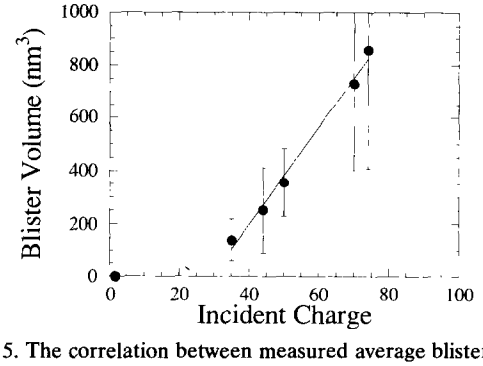

Fig. 5. The correlation between measured average blister type defect volumes and the incident charge of the ions (ions are Xe^{1+}, Kr^{35+}, Xe^{44+}, Xe^{50+}, U^{70+}, Th^{74+}). The error bars represent the standard deviation of the measured volume distributions.

9. Summary

The recently developed EBIT extracted ion facility is an effective tool in the investigation of various aspects of slow highly charged ion–solid surface interactions. The time scale of the transient hollow atom formation and decay to the ground state appears to be less than 100 fs, as indicated by X-ray emission and small angle scattered ion data. Evidence has been presented for the formation of a new ion induced surface defect, due to the electrostatic potential energy of the incident ion. A linear relationship between the defect volume and the incident ion charge supports a mechanism based on electron emission and subsequent collective displacements due to local charge imbalance.

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